

SUPPORT FOR THE AMENDMENTS

Claim 11 was previously canceled.

Claims 1 and 14 have been amended.

Claim 26-35 has been added.

The amendment of Claims 1 and 14 and the addition of new Claim 26-35 is supported by, for example, page 10, lines 16-20 of the specification as originally filed.

No new matter has been added by the present amendments.

REMARKS

Claims 1-10 and 12-35 are pending in the present application.

The rejections of:

- (a) Claims 1-6 and 9-13 under 35 U.S.C. §103(a) over Farrissey et al in view of Joern et al;
- (b) Claims 1-10, 12, and 13 under 35 U.S.C. §103(a) over Kohlstruck et al (US 2003/0187178) in view of Joern et al, and
- (c) Claims 14-25 under 35 U.S.C. §103(a) over Kohlstruck et al (US 2003/0187178) in view of Joern et al and Kohlstruck et al (US 6,093,817)

are respectfully traversed.

In the Office Action, the Examiner again openly recognizes that neither Farrissey et al nor Kohlstruck et al disclose or suggest “the presence of at least one trimerization catalyst selected from the group consisting essentially of the ammonium salts, substituted by four hydrocarbon radicals, of  $\alpha$ -hydroxy-carboxylates” according to Claim 1 of the present invention. However, the Examiner contends that Joern compensates for this deficiency by disclosing an alleged equivalence between  $\alpha$ -hydroxy-carboxylates and non- $\alpha$ -hydroxy-carboxylates (e.g., acetic acid which is disclosed by Farrissey et al and Kohlstruck et al) as trimerization catalysts. As such, the Examiner alleges that it would have been obvious to replace the trimerization catalysts used in Farrissey et al and Kohlstruck et al with an  $\alpha$ -hydroxy-carboxylate to provide “a process for preparing isocyanurate-containing polyisocyanates by at least partly trimerizing (cyclo)aliphatic diisocyanates, which comprises carrying out the reaction in the presence of at least one trimerization catalyst selected from

the group consisting essentially of the ammonium salts, substituted by four hydrocarbon radicals, of  $\alpha$ -hydroxy-carboxylates.”

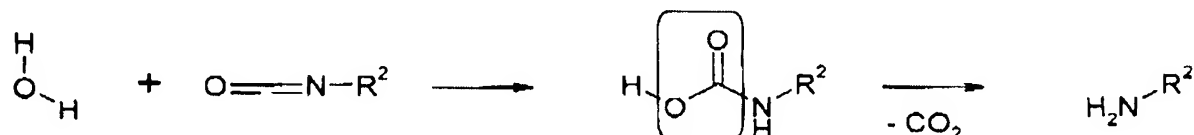
The Examiner also now cites a second Kohlstruck reference (US 6,093,817); however, this reference offers nothing further beyond that of the previously cited Kohlstruck reference. As such, Kohlstruck et al (US 6,093,817) also fails to disclose or suggest “the presence of at least one trimerization catalyst selected from the group consisting essentially of the ammonium salts, substituted by four hydrocarbon radicals, of  $\alpha$ -hydroxy-carboxylates” as required by Claim 14. Again, the Examiner’s contention is that Joern compensates for this deficiency by disclosing an alleged equivalence between  $\alpha$ -hydroxy-carboxylates and non- $\alpha$ -hydroxy-carboxylates and, thus, the claimed invention would be obvious.

Applicants disagree and submit that the Examiner’s interpretation of Joern is incorrect. Even if the artisan were to follow the argument that Joern discloses the combination of certain carboxylic acids (paragraphs [0009] and [0012]) with trimerization catalysts (paragraph [0013]), the catalysts disclosed by Joern are “tertiary amines, triazines, and, most preferably, metal salt trimerisation catalysts.” In other words, even if the skilled artisan were to select the tertiary amines from the list of trimerization catalysts and the alpha-hydroxy carboxylic acids from the list of carboxylic acids, the skilled artisan would not arrive at the claimed invention.

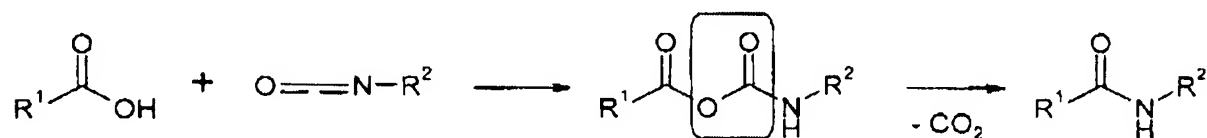
The examiner mainly refers to Joern in which a method for making urethane-modified polyisocyanurate foams is disclosed using

- a catalyst for trimerization (paragraphs [0013] to [0015]),
- a functionalized carboxylic acid (paragraphs [0011] to [0012]), and
- a blowing agent (paragraphs [0022] to [0027]).

However, it should be noted that water and carbon dioxide-evolving compounds act as blowing agents and form a foam, which is the goal in Joern. Water forms an unstable carbamic acid with an isocyanate ( $R^2$ -NCO) which evolves carbon dioxide and leaves the corresponding amine ( $R^2$ -NH<sub>2</sub>) as follows:



A free acid ( $R^1$ -COOH), however, acts as a blowing agent as it reacts with an isocyanate ( $R^2$ -NCO) under formation of an unstable anhydride which evolves carbon dioxide:



This reaction is of value in the reaction according to Joern, since a blowing agent for the formation of a foam is needed.

However, in the present invention the presence of carbon dioxide should be as little as possible, as carbon dioxide is known to act detrimental on the formation of isocyanurates from HDI, see CA 1335990 (copy **submitted herewith**; corresponds to EP 330966), in which the effect of a carbon dioxide content on ammonium hydroxide catalysts is taught, see also EP 524501 (copy **submitted herewith**).

The examples of CA 1335990 show that a higher content of carbon dioxide yields product with a high color number which is described to be "unsuitable for use in high-quality PUR [polyurethane] laquers", see CA 1335990, page 15. line 13.

EP 524501, at page 3. lines 49 to 52, discloses a pre-treatment of the isocyanates as preferred, in which the content of carbon dioxide can be reduced by bubbling an inert gas through the starting diisocyanate.

The main teaching from these two references is that the presence of carbon dioxide is to be avoided in the formation of isocyanurates for coating polyisocyanates, as the presence of carbon dioxide yields products with a high color number (see these two references and the comparative examples below). In polyurethane foams the color number is not relevant, since the foams usually not visible, e.g. inside seats. As water and free acids lead to the evolution of carbon dioxide they are also to be avoided for polyisocyanates for coatings.

Hence, the skilled artisan that wants to develop a process for the production of polyisocyanates for coatings would neither add water nor free acids to the catalyst and especially would not take into account the teaching of Joern, since it is known that the products obtained by such a process are not suitable for coatings. Thus, the prior art itself *teaches away* from the modification to the references that the Examiner alleges would be obvious.

Even if the skilled in the art would take into account the teaching of Joern, there is no expectation of success to receive products with a decreased color number.

It can easily be seen from page 8, lines 29-40 of the present specification how present invention copes with the presence of a free acid or water. More specifically, as detailed in this paragraph the catalyst according to the invention may be prepared by mixing a tetrasubstituted ammoniumhydroxide with a free acid. It is important that the water which is formed in this neutralisation is removed. e.g. by distillation. This is the reason why solvents forming an azeotrope with water are preferred.

In this way the formation of carbon dioxide by reaction of water with isocyanates can be suppressed.

Furthermore, the trimerization of the isocyanate is preferably carried out under an inert gas, see page 10, lines 16-20 of the present specification. This additional requirement is neither disclosed nor suggested by the cited art.

In addition, Applicants maintain for the reasons already of record that, contrary to the Examiner's allegations, there is simply no basis in the cited art to select the specific carboxylic acids of the present invention and specific quaternary ammonium salts of the present invention absent Applicants disclosure to serve as the guidepost. Further, as supported by the data in the specification, there is even less of a reasonable expectation created by the cited art of any level of success in combining their disclosures.

Indeed, Applicants maintain that data in the specification clearly illustrates that inclusion of an  $\alpha$ -hydroxy carboxylate provides for significant differences in the NCO values and the color number of the product obtained. These results are germane to the cited art for the reasons given above. Specifically, the cited art does not reasonable provide any basis for arriving at the claimed invention or any expectation of the benefits flowing therefrom. Accordingly, the data in the specification which shows the importance of the inclusion of an  $\alpha$ -hydroxy carboxylate speaks volumes as to the criticality of the method of the claimed invention.

Specifically, Applicants direct the Examiner's attention to the Examples of the present application, which clearly demonstrate that the  $\alpha$ -hydroxyl carboxylic acids give rise to polyisocyanates with a lower color number. Specific reference is made to the following comparisons:

Example 1 (color number of 30 Hz) vs. Comparative Example 1 (color number of 53 Hz)

Example 4 (color number of 17 Hz) vs. Comparative Example 2 (color number of 61 Hz)

Example 5 (color number of 180 Hz) vs. Comparative Example 3 (color number of 800 Hz)

Applicants submit that the results demonstrated in these comparisons clearly illustrate that, even if a *prima facie* case of obviousness can be established, Applicants demonstration is sufficient to rebut the same.

To further explain the relevance and importance of the foregoing comparison based on the data in the specification, Applicants submit the following additional remarks together with the following table specifically reporting the data from the examples of the present application.

	Catalyst	NCO-Value [%]	Color Number [Hz]
<b>Halogen-free hexamethylene diisocyanate (HDI)</b>			
Comp. Ex. 1	N-(2-hydroxypropyl)-N,N,N-trimethylammonium 2-ethylhexanoate	20.9	53
Ex. 1	carboxylate of tetramethylammonium hydroxide with 2-hydroxypropionic acid	19.0	30
Ex. 2	carboxylate of tetramethylammonium hydroxide with 2-ethyl-2-hydroxybutyric acid	21.2	21
Ex. 3	carboxylate of tetramethylammonium hydroxide with 2-hydroxyisocaproic acid	20.7	30
<b>HDI from phosgene process</b>			
Ex. 4	carboxylate of tetramethylammonium hydroxide with 2-ethyl-2-hydroxybutyric acid	21.2	17
Comp. Ex. 2	N-(2-hydroxypropyl)-N,N,N-trimethylammonium 2-ethylhexanoate	21.5	61

<b>Isophorone Diisocyanate</b>			
Comp. Ex. 3	N-(2-hydroxypropyl)-N,N,N-trimethylammonium 2-ethylhexanoate	17.2	800
Ex. 5	carboxylate of tetramethylammonium hydroxide with 2-ethyl-2-hydroxybutyric acid	17.1	180
Ex. 6	carboxylate of tetramethylammonium hydroxide with 2-hydroxypropionic acid	17.3	92

These data show that the color number is not affected by the NCO-value, see for example Examples 2 and 3 with higher and lower NCO-values than Comparative Example 1. The same is true for Examples 5 and 6 as compared to Comparative Example 3. The small differences in the NCO-values do not explain the differences in color number. In other words, the reason for the differences in the color number is not the NCO-value but the catalyst used in the examples.

Further, it is very difficult to exactly reproduce the NCO-values from one example to the other. As is evident the monomeric isocyanate is reacted in the presence of a catalyst to a certain NCO-value, depending on the yield, to obtain a reaction mixture comprising unreacted monomeric isocyanate and the reaction product. Accordingly, from the NCO-value of the reaction mixture the NCO-value of the product cannot necessarily be predicted.

Afterwards, the monomeric isocyanate is essentially distilled off, however, varying small amounts of the monomeric isocyanate remain in the product depending on the exact distillation conditions. This explains why the NCO-value can hardly be met even if you reproduce an example literally.

Looking at the comparison of comparative examples 1 and 2 to Examples 2 and 4, it can be seen that comparative examples 1 and 2 use the same catalyst by HDI of different



origin. Using a chlorine-containing HDI instead of a chlorine-free HDI yields a product which is slightly more colored. In contrast, using a catalyst according to the present invention and changing the isocyanate in the same way leads to a slightly less colored product of Example 4 compared to Example 2. This means that the catalysts according to the present invention are even more preferred in a process reacting a chlorine-containing HDI.

Again, Applicants submit that the data is germane to the cited art for the reasons that follow.

Looking at the disclosure of Joern et al, at paragraph [0027], lactic acid is known to act a blowing agent. However, in the manufacturing of polyisocyanates such a feature is not an advantage, but a disadvantage. Specifically, since the pure polyisocyanates are to be obtained rather than foams, the blowing agent feature is disadvantageous. Thus, based upon the disclosure of Joern et al the skilled artisan would not take into account lactic acid as part of a catalyst.

Looking a Kohlstruk et al, catalysts bearing a hydroxyl group are compared with catalysts not bearing a hydroxyl group. However, the hydroxyl group is part of the ammonium cation and not of the counterion, as in the present invention.

According to Kohlstruk et al, a catalyst bearing a hydroxyl group in the ammonium moiety (N-(2-hydroxypropyl)-N,N,N-trimethylammonium 2-ethylhexanoate (C-cat. 1), col. 9, lines 6-25) yield a product with a much higher color number than a catalyst with the same anion but not bearing a hydroxyl group (N,N-dimethyl-N-ethyl-N-(4-methoxybenzyl)ammonium 2-ethylhexanoate (cat. 4), col. 8, lines 43 et seq. and col. 9, lines 37-38). Accordingly, based on the disclosure of Kohlstruk et al, the skilled artisan would be led to the conclusion that adding hydroxyl groups to a catalyst is detrimental with respect to color number.

The Examiner attempts to disregard the foregoing evidence alleging at page 4 of the Advisory Action that “the fact that hydroxyl-substituted components was found to increase color number is

irrelevant to adding the hydroxy-substituent to the carboxylic acid component.” Aside from this nascent allegation, the Examiner makes no attempt to explain why the evidence of record is not being considered, much less explaining why the resultant benefit in the color number would be expected based on the cited art.

It is legal error for the Office to dismiss a showing of unexpected results as flowing from or inherent in the Examiner’s prior art construct (in this case, primarily the combination of Farrissey et al or Kohlstruck et al with Joern et al). As stated in In re Sullivan, 84 USPQ2d 1034 (Fed. Cir. 2007):

It is well settled that the PTO “bears the initial burden of presenting a prima facie case of unpatentability... . However, when a prima facie case is made, the burden shifts to the applicant to come forward with evidence and/or argument supporting patentability.” *In re Glaug*, 283 F.3d 1335, 1338 (Fed. Cir. 2002). Rebuttal evidence is “merely a showing of facts supporting the opposite conclusion.” *In re Piasecki*, 745 F.2d 1468, 1472 (Fed. Cir. 1984). Evidence rebutting a prima facie case of obviousness can include: “evidence of unexpected results,” *Pfizer, Inc. v. Apotex, Inc.*, 480 F.3d 1348, 1369 (Fed. Cir. 2007), evidence “that the prior art teaches away from the claimed invention in any material respect,” *In re Peterson*, 315 F.3d 1325, 1331 (Fed. Cir. 2003), and evidence of secondary considerations, such as commercial success and long-felt but unresolved needs, *WMS Gaming, Inc. v. Int’l Game Tech.*, 184 F.3d 1339, 1359 (Fed. Cir. 1999). When a patent applicant puts forth rebuttal evidence, the Board must consider that evidence. *See In re Soni*, 54 F.3d 746, 750 (Fed. Cir. 1995) (stating that “all evidence of nonobviousness must be considered when assessing patentability”); *In re Sernaker*, 702 F.2d 989, 996 (Fed. Cir. 1983) (“If, however, a patent applicant presents evidence relating to these secondary considerations, the board must always consider such evidence in connection with the determination of obviousness.”).

Rather than considering Applicants’ showing of unexpected results as rebuttal evidence to an alleged *prima facie* case, the Examiner has dismissed it. This is clear legal error.

Further, to illustrate the unexpected benefits of the claimed invention, the present inventors have prepared additional evidence of the effect of an increased amount of free acid in the reaction by preparing a series of reactions using different catalysts.

The catalysts were prepared by reaction of tetramethyl ammonium hydroxide with the appropriate acid. The acid was used

- in stoichiometric amounts with regard to the hydroxide ions,
- in a stoichiometric surplus of 10 mol%, and
- in a sub-stoichiometric amount of 10 mol%.

The carboxylic acid (lactic acid) was initially charged in a beaker and the tetrasubstituted ammonium hydroxide, dissolved as a 25% solution in methanol, was added under stirring. On completion of addition of the ammonium hydroxide, the mixture was stirred over night at room temperature. Subsequently, water of reaction formed and other volatile constituents were removed on a rotary evaporator at 90°C and 3 mbar. The residue was degassed and dissolved in 2-ethylhexanol as a 10% solution.

HDI was reacted to the isocyanurate trimer using these catalysts as described in the present specification. The reaction was conducted until a certain NCO value was reached and the reaction was stopped. The reaction mixture was distilled and the color number of the reaction mixture was determined before distillation and after distillation.

For comparative purposes DABCO® TMR was used as a catalyst, as in Comparative Example 1 in the present specification.

The results with tetramethylammonium lactate as catalyst are as follows:

	sub-stoichiometric amount of acid	stoichiometric amount of acid	stoichiometric surplus of acid	DABCO® TMR (comparative)
Reaction Time	25 min	35 min	55 min	65 min
NCO content	40.5%	40.5%	41.0%	41.6%
Color number (before distillation)	8 Hz	13 Hz	36 Hz	34 Hz
Color number (after distillation)	31 Hz	54 Hz	111 Hz	80 Hz
Amount of catalyst	202 ppm	204 ppm	230 ppm	265 ppm

It can easily be seen, that with the catalyst with a 10% stoichiometric surplus of the acid the reaction takes longer and needs more catalyst compared with the catalyst with a stoichiometric amount or a 10% sub-stoichiometric amount. However, the color number is higher and, therefore, not acceptable.

In contrast, the catalyst with the stoichiometric and sub-stoichiometric amount of acid provides products with a much better color number.

The inventor further prepared a series of examples in which the influence of acids on the reaction of HDI is examined.

For that purpose under nitrogen approx. 9 g HDI were placed into an ampoule, optional 1000 ppm acid and/or 500 ppm catalyst (benzyl trimethylammonium 2-hydroxy-isobutyrate) was added, and the reaction mixture was heated to 90°C for 60 min. A 2 ml syringe was put into the gas phase of the ampoule in order to compensate pressure, presumably due to carbon dioxide formation.

After that time the reaction was stopped with 2-hydroxyethyl carbamate and cooled down to room temperature. The NCO content and the color number of the reaction mixture was measured.

Example		NCO content (%)	Color number (Hz)	Vapor Compensation (ml)
1	Pure HDI	49.4	11	
2	+ 1000 ppm acetic acid	49.3	8	
3	+ 1000 ppm acetic acid + 500 ppm catalyst	37.6	161	0.8
4	+ 1000 ppm lactic acid	48.9	8	
5	+ 1000 ppm lactic acid + 500 ppm catalyst	36.8	189	2.0
6	+ 500 ppm catalyst	38.5	65	1.0

As can be seen from examples 2 and 4 no reaction takes place in the presence of acid. A reaction takes place only in the examples 3,5, and 6, in which catalyst is present. However, in those examples 3 and 5 in which the acid is present when the reaction takes place the color number deteriorates dramatically, regardless of the nature of the acid.

This demonstrates what was pointed out above, that the teaching of Joern et al. is not applicable for the formation of polyisocyanates for coatings: The presence of acids leads to products with a high color number.

In view of the foregoing, Applicants submit that the obviousness rejections are not sustainable and withdrawal of these grounds of rejection is requested.

Applicants submit that the present application is now in condition for allowance.

Early notification of such action is earnestly solicited.

Respectfully submitted,

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